

Final Report

**SURFACTANT-ENHANCED BICARBONATE FLOODING**

Project OE6, Milestone 7, Task 2B

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# SURFACTANT-ENHANCED BICARBONATE FLOODING

By Deborah A. Peru\*

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## FOREWORD

This final report concludes the third phase of the OE6 cooperative research project on "Surfactant-Enhanced Bicarbonate Flooding." This work was funded by the Department of Energy through cooperative agreement DE-FC22-83F360149 and Kerr-McGee Chemical Corporation through contract no. 85-132B. The work plan consisted of six tasks. An interim report, NIPER-181, was submitted to the Department of Energy and Kerr-McGee Chemical Corporation in August 1986. In evaluating the progress of this research project, we would recommend that both reports be read and evaluated together.

## ABSTRACT

Coalescence rate constants were calculated for systems containing alcohol ethoxysulfate both with and without Tronacarb™ (sodium bicarbonate). All systems containing Tronacarb above 3.8 percent total salinity had higher coalescence rate constants than systems not containing Tronacarb. The results indicate that Tronacarb promotes faster coalescence of crude oil-surfactant brine emulsions.

Addition of Tronacarb to preflush brine was found to be more economical than chloride brine in reducing divalent ion concentration. Silicon (Si) concentrations did not exceed 24 ppm after Tronacarb was in contact with Berea sandstone for 1 week at 42° C. The occurrence of silica scales is expected to be minimal when using Tronacarb in chemical flooding or in a preflush.

A chemical slug containing Tronacarb, petroleum sulfonate, and polymer recovered from 6 to 20 percent more residual oil than did systems containing either: Tronacarb plus polymer or surfactant plus polymer. The results from the oil-displacement tests indicate that a synergistic relationship exists between Tronacarb and low concentrations of surfactant and polymer whereby the oil-recovery efficiency was improved, and the chemical cost per barrel of oil recovered was decreased when the three chemicals were used together.

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## INTRODUCTION

A reduction in interfacial tension (IFT) between entrapped residual oil and chemical flooding fluids is a necessary condition for successful recovery of residual crude oil. Depending upon the ionic and chemical environment, a reduction in IFT between crude oil and chemical fluids may lead to the formation of stable emulsions. The coalescence of oil-brine macroemulsions is a critical phenomenon in oil recovery by chemical flood methods. Wasan, et al.<sup>1</sup> have found that certain additives which lower interfacial viscosity will promote the coalescence of oil droplets and enhance the formation of an oil bank.

The rate of coalescence is controlled by factors that stabilize the oil-brine emulsions. In alkaline flooding, the major cause of stabilized emulsions is the accumulation of interfacially active asphaltic aggregates at the oil-brine interface.<sup>2</sup> Inorganic bases, such as sodium bicarbonate or carbonate, and certain surfactants may break down the rigid films at the oil-brine interface to form liquid films.<sup>1-3</sup> The result is a system that has a greater tendency to coalesce.

The addition of synthetic surfactant to sodium bicarbonate has been shown to cause an immediate reduction in interfacial tension.<sup>4</sup> In oil-displacement tests, a chemical formulation that lowers IFT quickly should recover the most oil because of the faster rate of oil emulsification. While a reduction in IFT may reduce the energy required for forming emulsions, it may also increase the energy required for breaking them.

In this study, the coalescence of emulsions formed with synthetic surfactants was compared with the coalescence of emulsions formed with sodium bicarbonate and synthetic surfactant (task 1b, NIPER Proposal No. 85-132B). The initial rate of coalescence was observed by measuring the rate of water-breakout in phase tubes after creaming (emulsification followed by phase separation). The effect of adding sodium bicarbonate to synthetic surfactant was evaluated by comparing the coalescence rate constants of these systems with the coalescence rate constants of systems using synthetic surfactants alone.

The advantages of using Tronacarb (sodium bicarbonate) as an alkaline additive in chemical flooding include (1) the ability to reduce surfactant adsorption, (2) alter wettability, (3) to maintain low interfacial viscosity, and (4) to reduce divalent ion concentrations. However, a major advantage of using sodium bicarbonate as an alkaline flooding agent, is that it does not react with most reservoir minerals including quartz. The dissolution of quartz can cause deposition of silica scales that are almost impossible to remove from producing wells. It is critical that the formation and deposition of silica scales be minimized during an alkaline chemical flood, because wellbore scale inhibitors are not very effective.<sup>5</sup>

In this study, the use of a Tronacarb solution as a preflush was compared with a sodium chloride solution for the ability to reduce divalent ion concentrations to levels tolerated by low concentrations of surfactant. The amount of silicon in the effluent after 1 week of contact with Berea sandstone was determined and compared with literature results for the stronger alkaline agents (task 3). This experiment was performed as an alternative to the interfacial viscosity measurements because it was more important to establish that silica dissolution was minimized using sodium bicarbonate.

The final task (task 6) in this EOR research project was to determine the amount of residual oil recovered by the most favorable surfactant-enhanced Tronacarb processes. Determination of a synergistic effect between Tronacarb and low concentrations of surfactant and polymer was established by performing two control oil-displacement tests.

## EXPERIMENTAL

Coalescence studies were performed on the same systems used in the emulsion screening tests. The systems contained Wilmington crude oil and aqueous chemicals at a WOR of 1. Brines contained either 0.1 percent ethoxysulfate and NaCl or 0.1 percent ethoxysulfate, 1.2 percent NaHCO<sub>3</sub> (Tronacarb) + NaCl at various total salinities. Emulsions were formed by using a hand homogenizer. The coalescence of the emulsions was observed by placing the phase tubes vertically in a 70° C oven and monitoring the breakout of brine over time.

The preflush experiments were performed using two 10-in. unfired consolidated Berea sandstone cores. Each core was saturated and equilibrated

with 3 percent NaCl and 1,000 ppm each Ca and Mg ions. Three fourths of a pore volume of either 3 percent NaCl or 2.2 percent NaHCO<sub>3</sub> + 1.5 percent NaCl was injected and the cores were shut-in for 1 week at 42° C; then flow was resumed and the effluent was analyzed for Ca and Mg. During the postflood procedure, the pressure drop across the core was monitored to detect any permeability loss caused by plugging. No change in pressure drop across the core was observed during this experiment.

When the experiment was completed, the cores were cleaned with 50 percent aqueous isopropyl alcohol solution, 0.01 N HCl solution, and were resaturated and equilibrated with 3 percent NaCl and 1,000 ppm each Ca and Mg ions. The experiment was repeated at 49° C using opposite cores for preflush chemical injection. There was no shut-in period during the second experiment; that is, the chemicals were continuously injected. Ca and Mg ion concentrations were analyzed with an atomic absorption spectrophotometer. Silicon was determined by forming a yellow complex with molybdic acid and measuring the absorbance at 450 nm with a Beckman spectrophotometer.<sup>6</sup> Alkalinity was measured by titration with standardized 0.1 N HCl to a methyl red endpoint (pH 4.8).

Oil-displacement tests were performed at a linear advance rate of 1 ft/d. Hassler core holders were maintained at 50° C in all experiments using heated thermal jackets. Overburden pressure was maintained at > 200 psi. The viscosities of all chemical slug solutions were measured with a Brookfield LVT cone/plate viscometer to ensure that all graded polymer solutions were of the same viscosity ±2 cp. The polymer used in these experiments was Flocon<sup>®</sup> 4800C (12.5 percent active) obtained from Pfizer Chemical company. The surfactants used were Neodol<sup>®</sup> 25-3S alcohol ethoxysulfate (58.8 percent active) obtained from Shell Chemical Co. and TRS 10-410 petroleum sulfonate (62 percent active) obtained from Witco Chemical Co., Sonneborn Division. Tronacarb, sodium bicarbonate (94 percent active), was obtained from Kerr-McGee Chemical Co.

## RESULTS

### Coalescence Studies

The effect of two chemical additives on the coalescence of Wilmington crude oil-brine emulsions was observed by monitoring the rate of brine coalescence. A method of fitting the observed coalescence data has been developed and is used to calculate the coalescence rate constants and the plateau levels from the fit parameters. The rate equation used is:  $y = a(1 - e^{-kt})$ ; where  $y$  is the volume of lower phase that has coalesced per total volume of emulsion,  $a$  is the plateau value,  $k$  is the coalescence rate constant, and  $t$  is time.

Figures 1 through 5 show both the fitted coalescence curve (solid lines) and the observed values (+) for systems containing synthetic surfactant alone or synthetic surfactant plus Tronacarb. Good agreement between the fitted coalescence curves and the observed values is shown for all salinities tested with the exception of 5.33 percent.

Table 1 lists the calculated values for the plateau level ( $a$ ) and the coalescence rate constant ( $k$ ) for the 10 systems studied. Comparison of the ( $k$ ) and ( $a$ ) values for systems both with and without Tronacarb shows that although the systems without Tronacarb are unaffected by salinity, the Tronacarb systems are very sensitive to added sodium chloride. At low salinity, the initial rates of coalescence ( $k$ ) for emulsions with and without Tronacarb are similar but Tronacarb reduced the amount of stable emulsion (higher plateau level). At higher salinities, Tronacarb increased the coalescence rate constant and the amount of stable (irreducible) emulsion. Sodium chloride by itself reduced the amount of stable crude oil-surfactant emulsion, but its effect on rate failed to show a definite trend.

The coalescence rate constants can be used with interfacial tension data<sup>4</sup>, and interfacial viscosity data (to be obtained in the future) and correlated to oil-displacement results. Systems that have fast rates of coalescence are expected to have low interfacial viscosities and are more likely to coalesce to form an oil bank causing a greater reduction in the residual oil saturation.

## Linear Flow Preflush Experiment

The purpose of this experiment was to determine whether a detectable difference in the resident brine divalent ion concentration would occur if a bicarbonate solution or a chloride solution were injected into two separate Berea cores pre-equilibrated with 1,000 ppm each of calcium and magnesium ions.

Figure 6 shows that a detectable difference does occur between a Tronacarb (bicarbonate) preflush and a chloride brine preflush. The larger area under the magnesium and calcium curves in the chloride experiment implies that divalent ions are precipitated during the shut-in period using bicarbonate. In general, there is a 1.0-pore volume lag in the reduction in divalent ion concentration using chloride brine. At a frontal advance rate of 1 ft/d, an additional 1.3 years would be required to preflush a 10-acre pattern using chloride brine than a preflush using Tronacarb brine.

Figure 7 shows the concentration of silicon in the effluent after sodium bicarbonate (2.2 percent) or sodium chloride (3.0 percent) solutions were in contact with Berea sandstone for 1 week. Shortly after 1 pore volume of fluid was injected, the concentration of silicon rose to 24 ppm in the bicarbonate solution and to 6 ppm in the chloride solution. Alkalinity in the effluent was also measured to determine breakthrough of the bicarbonate solution. Bicarbonate broke through at 1 pore volume and quickly rose to the injected concentration.

Table 2 lists the silicon concentrations in effluents from corefloods using stronger alkaline solutions.<sup>7</sup> The higher level of silicon in the effluent at higher pH indicates a much greater reactivity with quartz.<sup>8</sup> Application of the Tronacarb process in the reservoir will minimize quartz dissolution, so that silicon concentrations will not exceed 24 ppm.

To ensure that the difference in the divalent ion concentration in the effluent was not a result of a variation in core pore size distribution, the experiment was repeated. The core previously injected with chloride solution was now injected with Tronacarb solution and vice versa. The chemicals were continuously injected (no shut-in period), and the effluent was analyzed for divalent ion concentration as before.

Table 3 lists the concentration of divalent ions flushed from the Berea sandstone in equilibrium with the divalent ions. The average weight of calcium displaced by the Tronacarb preflush was 29 percent lower than when using the chloride preflush. Likewise, the weight of magnesium displaced from each core was an average of 11 percent lower using the Tronacarb preflush solution than when using the chloride preflush solution. Reproducibility of the results obtained in the two trials was greater than 90 percent.

These results indicate that addition of Tronacarb to preflush waters is more effective at reducing divalent ion concentration than use of sodium chloride brine. The reduced concentration of divalent ions in the resident brine suggests that bicarbonate may be precipitating calcium as calcium carbonate and magnesium as magnesium silicate, thereby reducing their concentrations in the resident brine. This will reduce the amount of surfactant-divalent ion precipitation during the injection of slugs containing low concentrations of surfactant.

#### Oil Displacement Tests

Table 4 lists the oil recovery efficiency and the chemical cost per barrel of oil produced using the surfactant-enhanced bicarbonate systems found to be most promising in the emulsion screening tests. A Tronacarb solution enhanced with petroleum sulfonate recovered more residual oil than did the Tronacarb solution enhanced with alcohol ethoxysulfate. This additional recovery could result from the lower equilibrium interfacial tension obtained with the former system<sup>4</sup> or the difference in the injection strategy, as shown in figures 8 and 9.

Figure 8 shows the oil cut produced versus cumulative pore volume of injected fluid for the system containing Tronacarb, ethoxysulfate and polymer. In this experiment, a high concentration of surfactant was injected first, followed by three graded polymer slugs. All chemical slugs contained 1.2 active percent Tronacarb and 8.83 percent sodium chloride (pH 9). The average oil recovery efficiency was 24 percent, and the approximate chemical cost per barrel of oil produced was \$20 including the cost of polymer. Figure 9 shows the oil recovery results for the Tronacarb system enhanced with petroleum sulfonate. In this experiment, 0.2 percent of petroleum sulfonate was injected into the core for approximately 1 pore volume. The surfactant

slugs contained 1.2 active percent Tronacarb and 0.97 percent sodium chloride with decreasing amounts of polymer (pH 9). The average oil recovery efficiency was 32 percent, and the average chemical cost per barrel of oil produced was \$14 including the cost of polymer.

Since the Tronacarb solution enhanced with petroleum sulfonate recovered the most residual oil, it was chosen as the system to be used in performing control studies. One control test was performed using petroleum sulfonate and polymer without added Tronacarb (pH 5), and the other control was run using Tronacarb and polymer without surfactant (pH 9). The data in table 4 show that neither of the controls recovered as much Wilmington crude oil as did the three chemicals together. The results indicate that Tronacarb enhanced with low concentrations of surfactant and polymer can recover from 6 to 20 percent more residual oil than either of the two components alone. A synergistic relationship appears to exist, as it did in the IFT experiments, between Tronacarb and low concentrations of surfactant (and polymer).

Shown in table 5 are the individual chemical costs per barrel of oil produced during a surfactant-enhanced Tronacarb flood with polymer. In all cases, polymer was the major chemical cost. In enhanced oil recovery, the chemical cost for implementing a surfactant-enhanced Tronacarb process with added polymer can be reduced by decreasing the concentration of bio-polymer or by using a synthetic (low cost) polymer whenever possible.

In secondary waterflooding, polymers are sometimes used to accelerate the recovery of oil by providing better mobility control. Although polymers can improve sweep efficiency they do not displace the trapped residual oil droplets that remain in the rock pore spaces. At a low cost, Tronacarb could also be added to a polymer-augmented waterflood which would expedite oil recovery by mechanisms that normally would not be encountered using polymer alone, such as, reduced interfacial tension, lower interfacial viscosity, emulsification and wettability changes. Research at NIPER has shown that Tronacarb (sodium bicarbonate) has the potential for improving oil recovery during both secondary and tertiary stages of oil production.

## EXPERIMENTAL CONCLUSIONS

1. The exponential equation,  $y = a(1 - e^{-kt})$ , fits the observed coalescence data and is used to find the coalescence rate constants for Wilmington crude oil-brine macroemulsions.
2. Addition of Tronacarb to alcohol ethoxysulfate increases the rate constant for surfactant-brine coalescence for the intermediate to high-salinity brines (5.3 to 12.8 percent). At low salinity (3.8 percent), systems with and without Tronacarb have similar coalescence rate constants.
3. Addition of Tronacarb to preflush waters is economically more effective than plain brine in reducing the concentration of divalent ions in the resident brine. Divalent ion precipitates will be less likely to affect surfactant concentration than will divalent ions in solution.
4. The low concentration of silicon in effluents from corefloods using the "Tronacarb process" will minimize silica scale formation.
5. A chemical slug containing petroleum sulfonate, Tronacarb, and polymer recovers more residual oil than a system containing alcohol ethoxysulfate, Tronacarb, and polymer. Control experiments using two-component chemical slugs indicate that both the chemical cost per barrel of oil recovered and the oil recovery efficiency are improved when surfactant, Tronacarb, and polymer are used together.

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TABLE 1. - Values for a and k at various salinities with and without Tronacarb™. The coalescence rate equation is:  $y = a(1 - e^{-kt})$ ; where t is time and y is the fraction of total volume for lower phase coalescence

NaCl added to Neodol® ethoxysulfate systems, %	a is the plateau level (extent of coalescence)		k is the coalescence rate constant	
	NaCl <u>alone</u>	with <u>NaHCO<sub>3</sub></u> <sup>1</sup>	NaCl <u>alone</u>	with <u>NaHCO<sub>3</sub></u> <sup>1</sup>
3.83	0.409	0.457	0.103	0.101
5.33	0.426	0.525	0.097	0.145
6.83	0.469	0.524	0.132	0.216
10.83	0.494	0.480	0.148	0.304
12.83	0.505	0.419	0.130	0.514

<sup>1</sup> 1.2% NaHCO<sub>3</sub> (Tronacarb™).

TABLE 2. - Silicon concentration in produced fluids from alkaline Berea corefloods<sup>1</sup>

Alkali	Wt. % Na <sub>2</sub> O	Peak Si levels, ppm
NaHCO <sub>3</sub>	<sup>2</sup> 0.44	<sup>2</sup> 24
Na <sub>2</sub> CO <sub>3</sub>	0.50	33
Na <sub>2</sub> CO <sub>3</sub>	1.00	47
NaOH	0.50	210
NaOH	1.00	420
Na <sub>4</sub> SiO <sub>4</sub>	0.50	999 <sup>3</sup> (1,132)
Na <sub>4</sub> SiO <sub>4</sub>	1.00	2,548 <sup>3</sup> (2,263)

<sup>1</sup>Reference 7, Burke, J. H. SPE paper 12039.

<sup>2</sup>This study.

<sup>3</sup>Injected Si levels in parentheses.

TABLE 3. - Concentration of divalent ions flushed out per volume of rock pore space. Temperature was maintained at 45° C for trial 1 and 49° C for trial 2

Preflush	Magnesium, mg/cm <sup>3</sup>		Calcium, mg/cm <sup>3</sup>	
	Trial 1	Trial <sup>1</sup> 2	Trial 1	Trial <sup>1</sup> 2
2.2% NaHCO <sub>3</sub> (Tronacarb)	1.02	1.10	0.79	0.82
1.5% NaCl				
3.0% NaCl	1.13	1.26	1.17	1.10

<sup>1</sup>Cores were reversed, presaturated with divalent ions, and preflushed using Tronacarb™ and chloride or chloride alone. The preflush chemicals were not shut in. 1.9 pore volumes of each chemical were continuously injected.

TABLE 4. - Oil displacement at 50° C for surfactant-enhanced sodium bicarbonate floods of Wilmington crude oil (viscosity = 65 cp at 50° C)

Process	Trial #	S <sub>orw</sub>	S <sub>orc</sub>	R.E. <sup>1</sup> , %	Chemical cost, \$ per barrel of oil produced
<u>Surfactant-enhanced bicarbonate plus polymer</u>					
1/4 PV - 0.65% alcohol ethoxylate, 1.2% NaHCO <sub>3</sub> , 8.83% NaCl, and 58 ppm total polymer followed by 3/4 PV of 1,100 ppm total polymer (graded as shown in figure 8). This was followed by 1 PV of a 8.83% NaCl water drive.	1	43.5	33.4	23.2	20
	2	38.8	33.9	<sup>2</sup> 12.6	<sup>2</sup> 42
1 PV - 0.2% petroleum sulfonate, 1.2% NaHCO <sub>3</sub> , 0.97% NaCl, and 1,100 ppm total polymer (graded as shown in figure 9). This was followed by 1 PV of a 1.8% NaCl water drive	3	48.6	32.6	33.1	12
	4	43.2	29.9	30.8	15
<u>Surfactant plus polymer</u>					
1 PV - 0.2% petroleum sulfonate + 1.8% NaCl with 1,100 ppm total polymer graded followed by 1 PV of a 1.8% NaCl water drive	5	43.5	32.1	26.2	14
<u>Bicarbonate plus polymer</u>					
1 PV - 1.2% NaHCO <sub>3</sub> + 0.97% NaCl with 1,100 ppm total polymer graded followed by 1 PV of a 1.8% NaCl water drive	6	41.4	37.1	10.4	26

$$^1 \text{Recovery efficiency} = \frac{S_{orw} - S_{orc}}{S_{orw}} \times 100$$

<sup>2</sup>These results are probably inaccurate because the oil saturated core was not aged more than 24 hours.

TABLE 5. - Individual chemical cost per barrel of oil recovered  
based on laboratory oil displacement test results

Trial #	Polymer Cost, \$ per barrel of oil recovered	Surfactant Cost, \$ per barrel of oil recovered	Tronacarb Cost, \$ per barrel of oil recovered	Total Chemical Cost, \$ per barrel of oil recovered
1	11	4	5	20
3	6	4	2	12
4	8	5	2	15

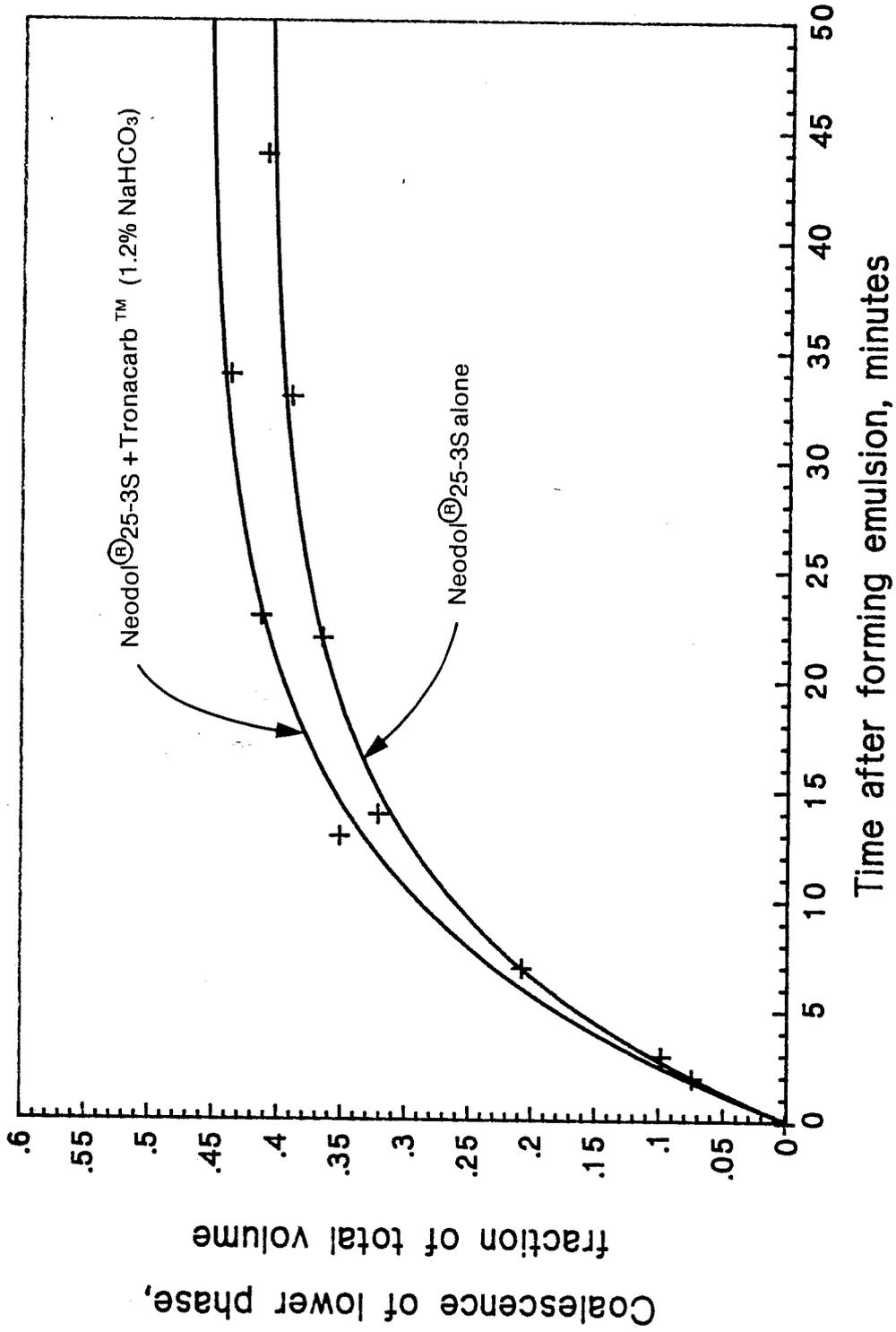


Figure 1. - Coalescence of the lower phase versus time. Systems contain Wilmington crude oil and 0.1 percent alcohol ethoxysulfate plus 3.83 percent sodium chloride alone or combined with 1.2 active percent Tronacarb. WOR = 1.

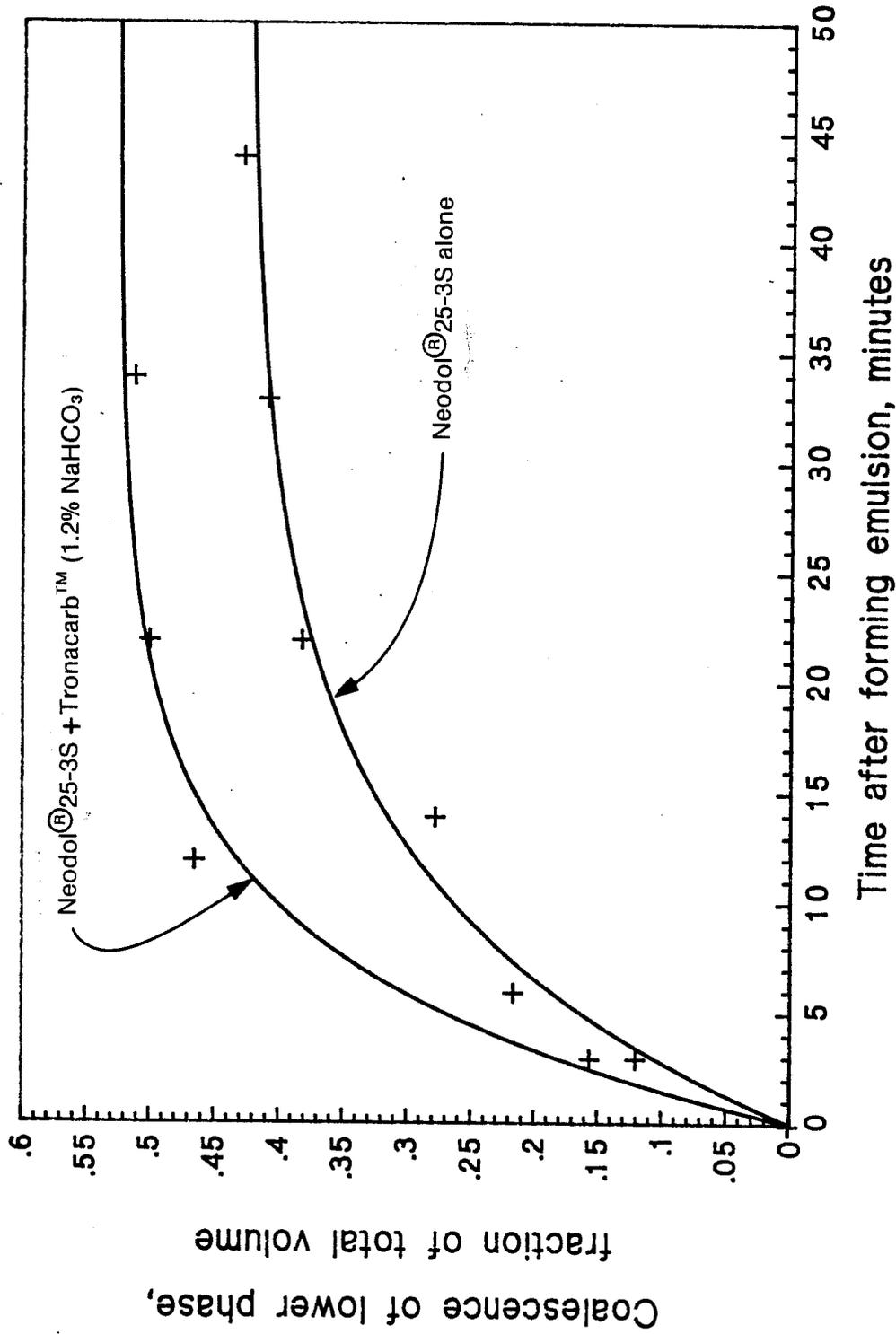


Figure 2. - Coalescence of the lower phase versus time. Systems contain Wilmington crude oil and 0.1 percent alcohol ethoxysulfate plus 5.33 percent sodium chloride alone or combined with 1.2 active percent Tronacarb. WOR = 1.

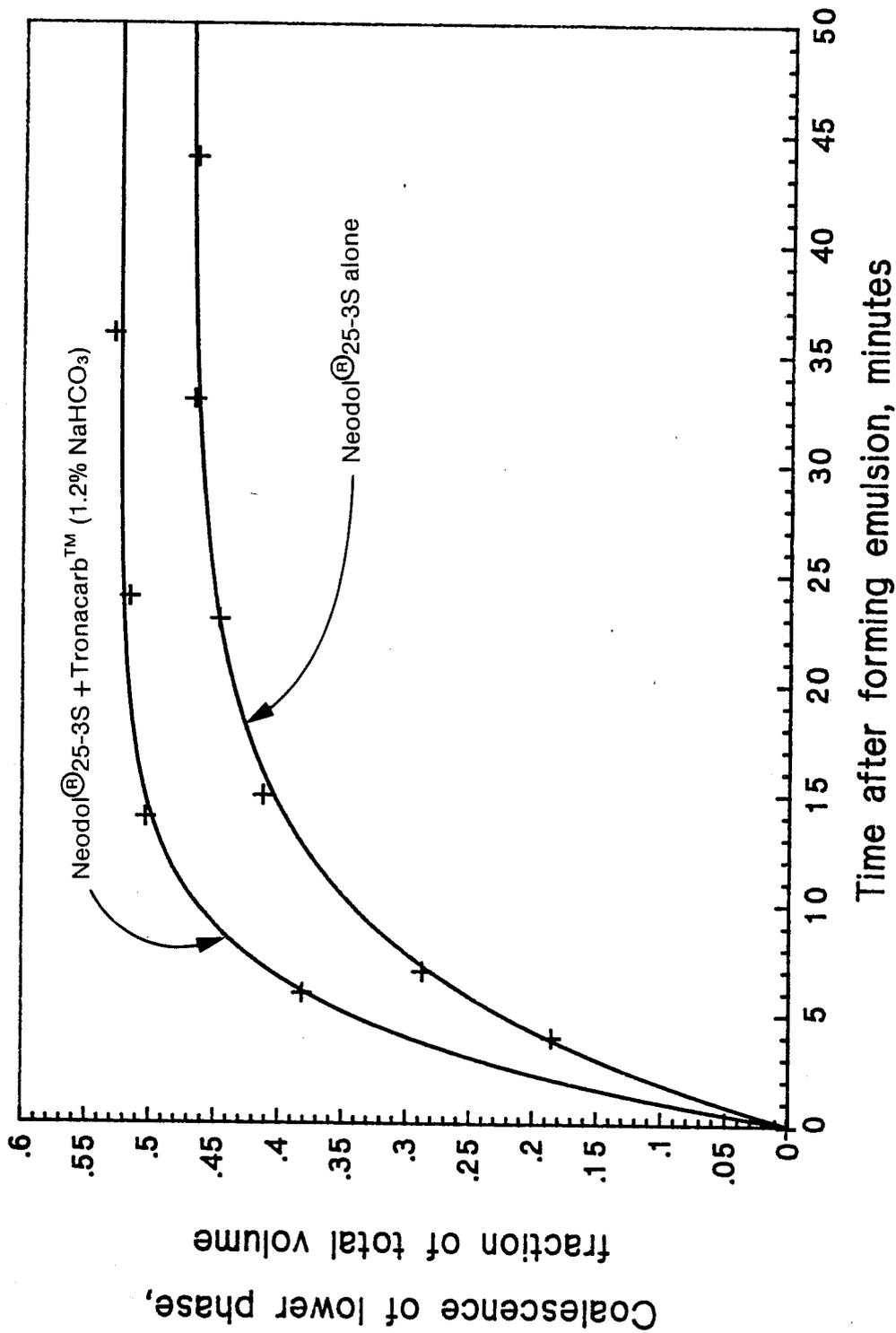


Figure 3. - Coalescence of the lower phase versus time. Systems contain Wilmington crude oil and 0.1 percent alcohol ethoxysulfate plus 6.83 percent sodium chloride alone or combined with 1.2 active percent Tronacarb. WOR = 1.

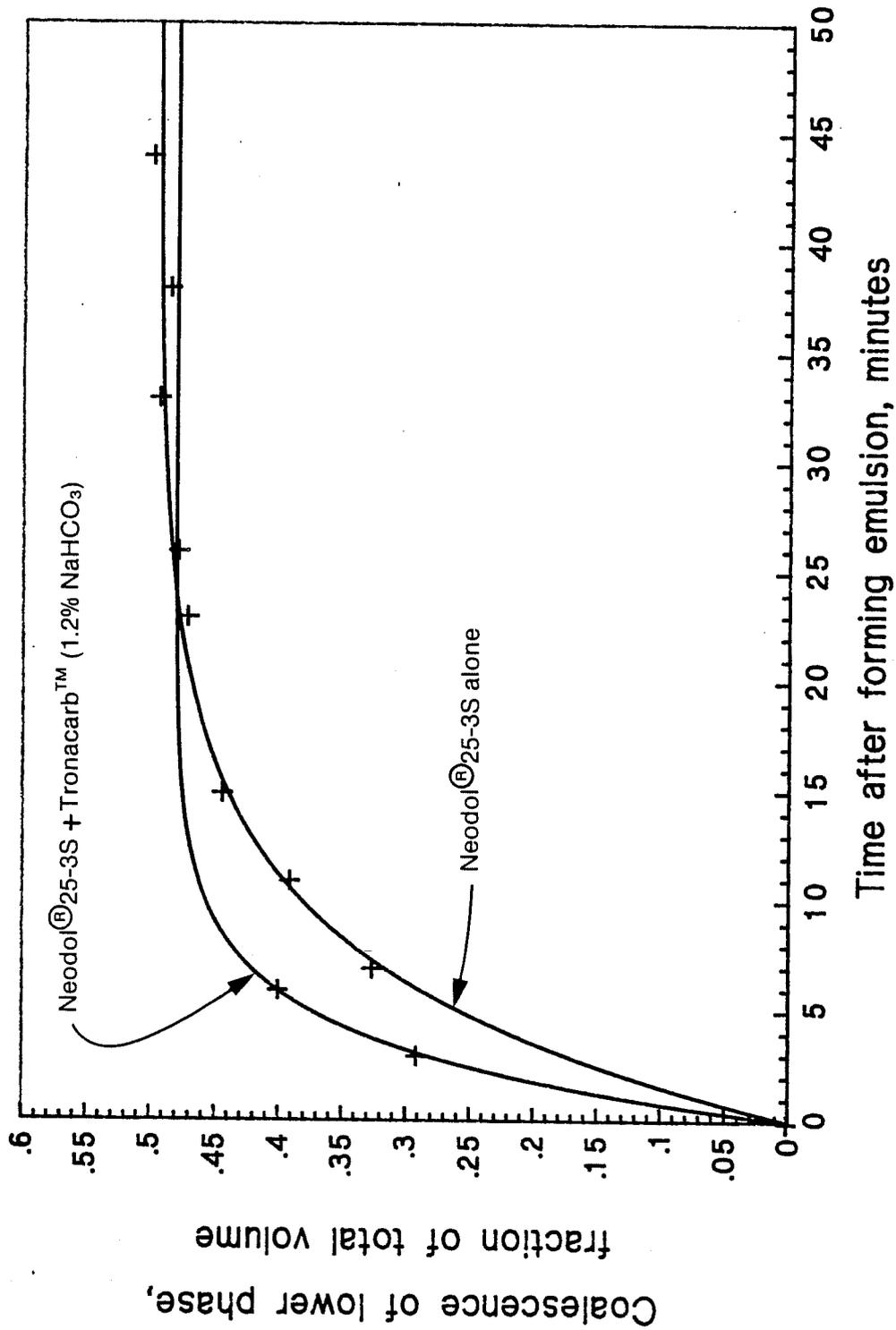


Figure 4. - Coalescence of the lower phase versus time. Systems contain Wilmington crude oil and 0.1 percent alcohol ethoxysulfate plus 10.83 percent sodium chloride alone or combined with 1.2 active percent Tronacarb. WOR = 1.

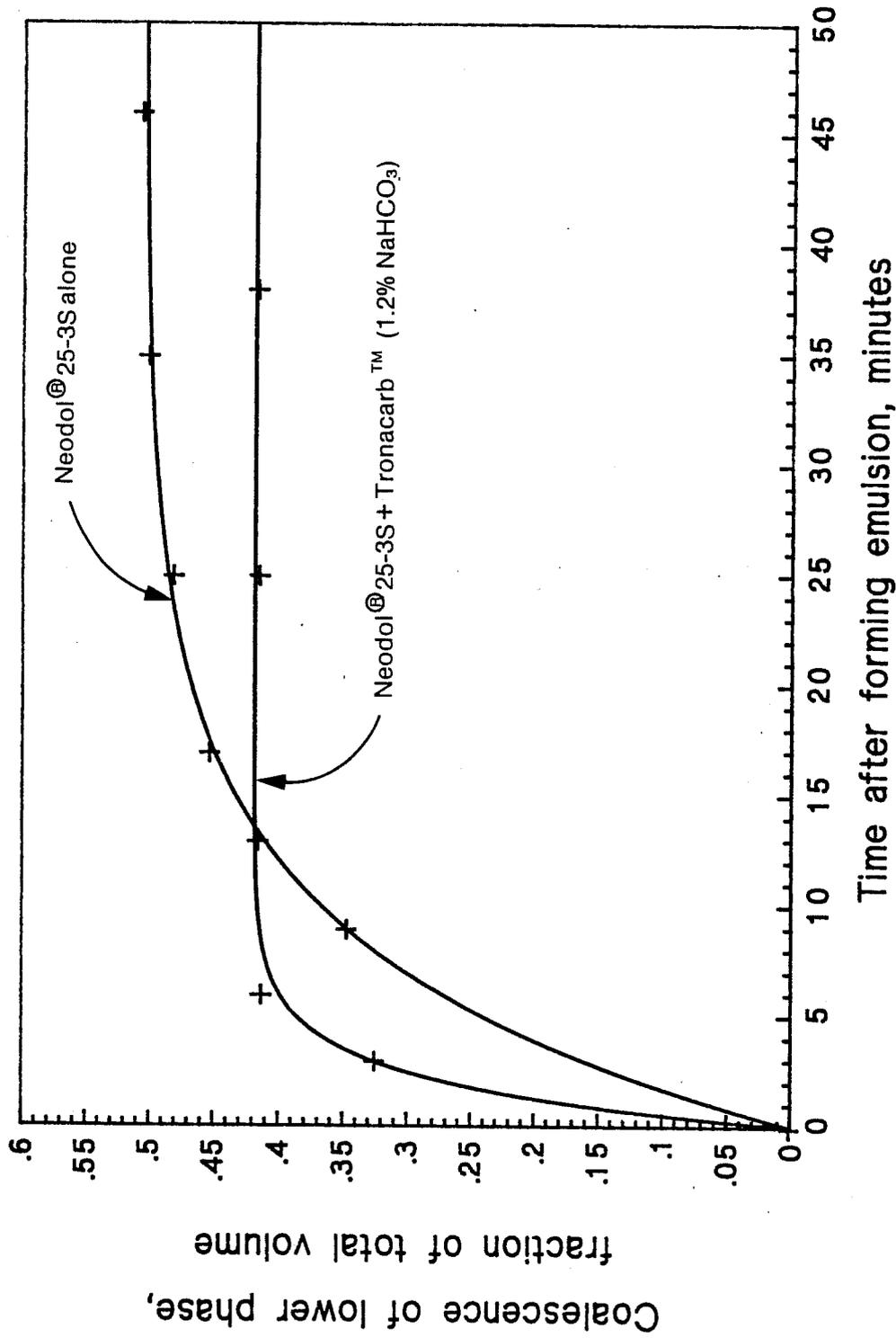


Figure 5. - Coalescence of the lower phase versus time. Systems contain Wilmington crude oil and 0.1 percent alcohol ethoxysulfate plus 12.83 percent sodium chloride alone or combined with 1.2 active percent Tronacarb. WOR = 1.

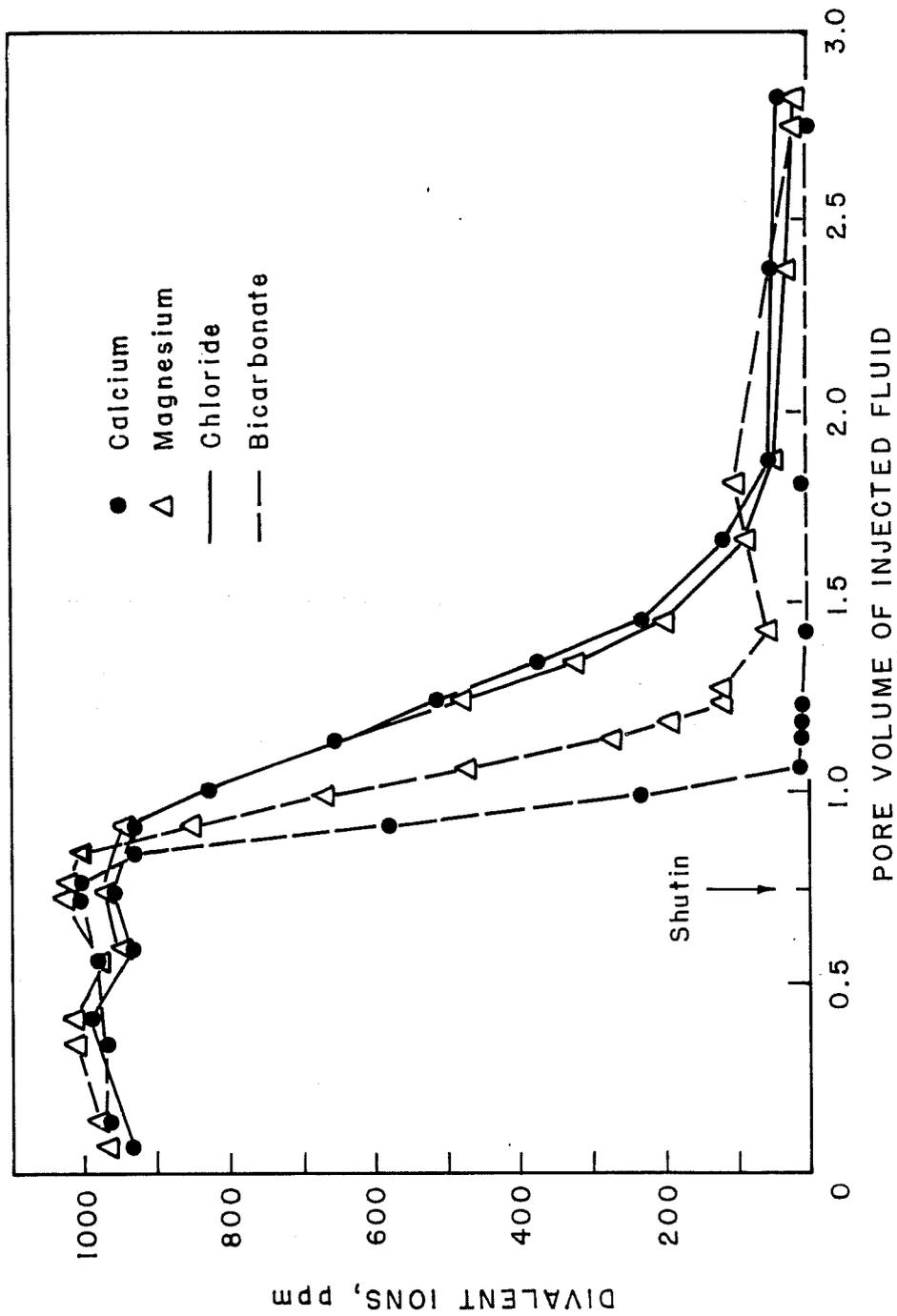


Figure 6. - Plot of divalent ion concentration in effluent after bicarbonate or chloride solutions were shut into Berea sandstone cores for 1 week at 42° C. Effluent was analyzed after flow was resumed.

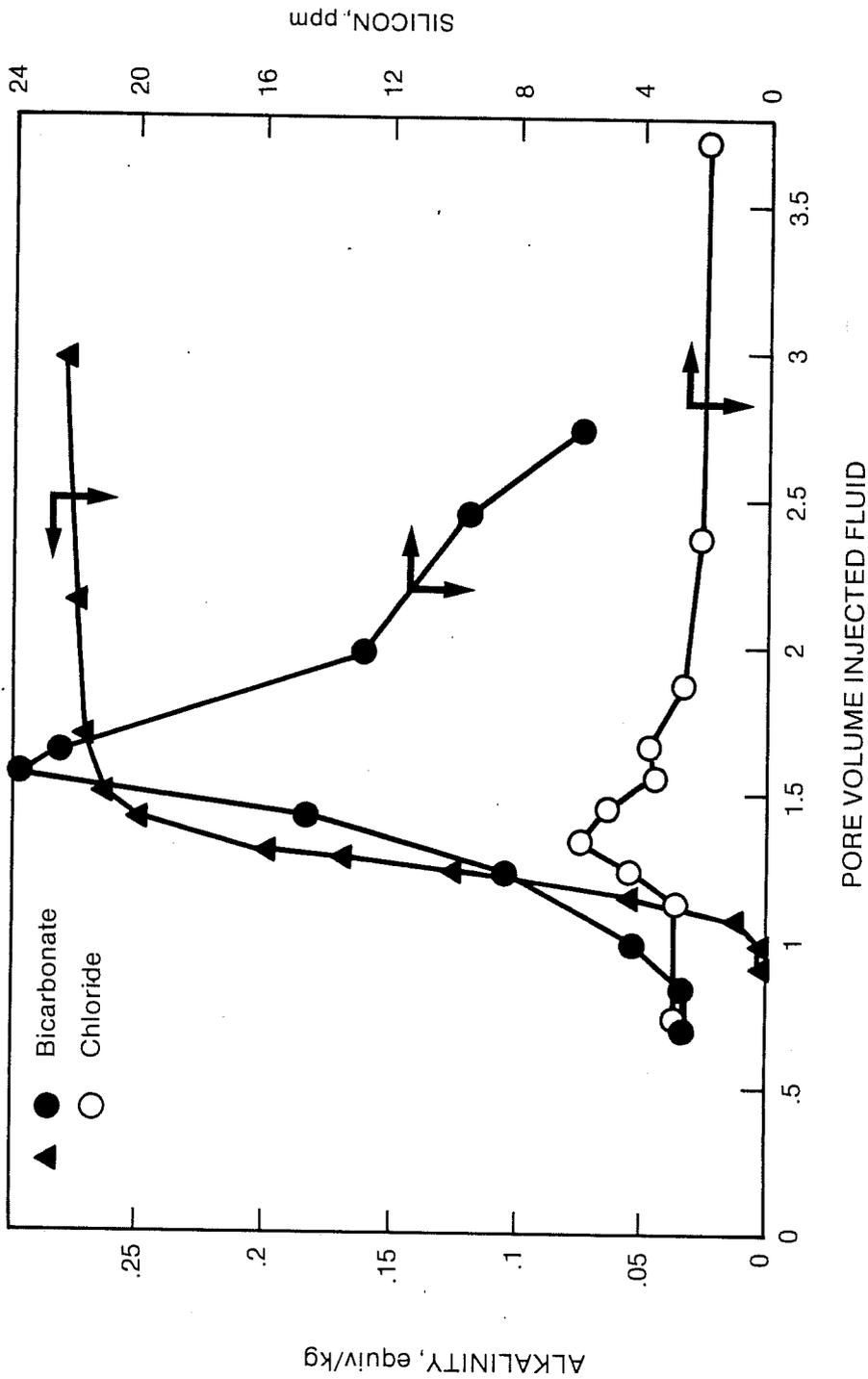


Figure 7. - Plot of silicon concentration in the effluent after bicarbonate or chloride solutions were in contact with Berea sandstone cores for 1 week at 42° C. The effluent from the bicarbonate preflush was analyzed for total alkalinity to determine breakthrough of bicarbonate.

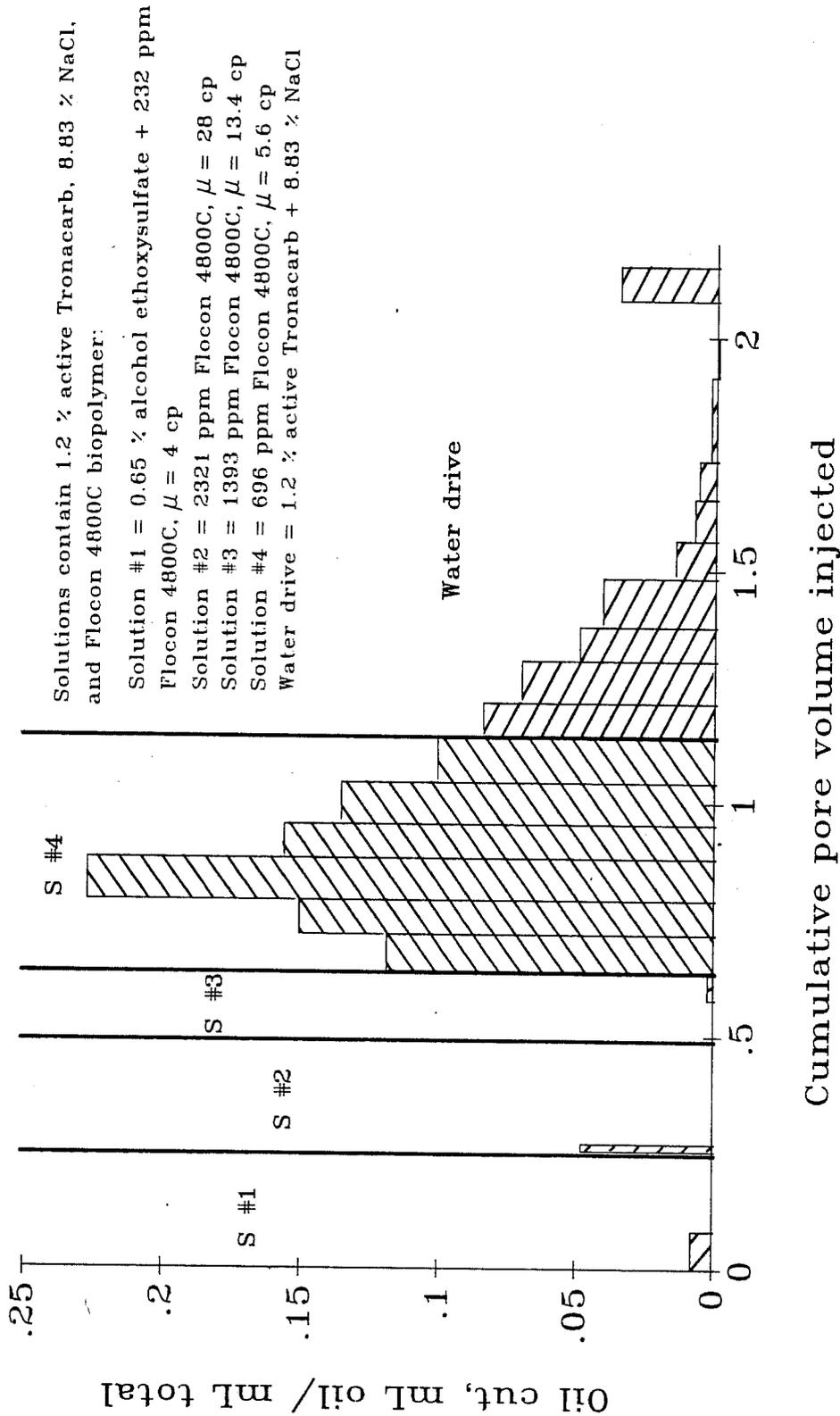


Figure 8. - Shell Neodol 25-3S-enhanced Tronacarb flood. Plot shows oil cut versus cumulative pore volume of tertiary chemical injected into Berea sandstone initially at residual oil saturation. Viscosity of Wilmington crude oil = 65 cp at 50° C.

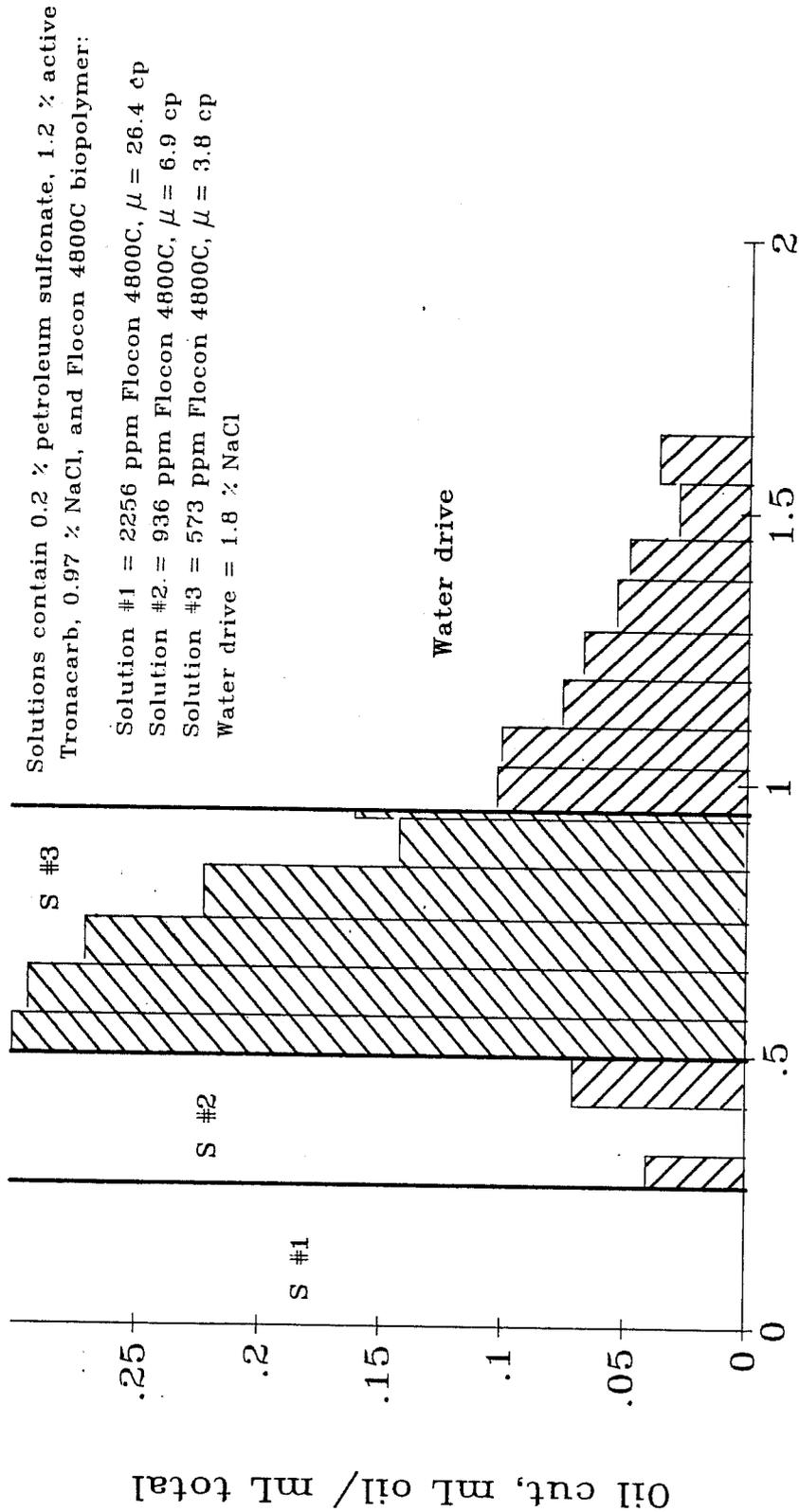


Figure 9. - Witco TRS 10-410-enhanced Tronacarb flood. Plot shows oil cut versus cumulative pore volume of tertiary chemical injected into Berea sandstone initially at residual oil saturation. Viscosity of Wilmington crude oil = 65 cp at 50° C.